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# IDENTIFICATION AND YIELD OF $Pd^{117}$ IN THE THERMAL-NEUTRON FISSION OF $U^{235}$

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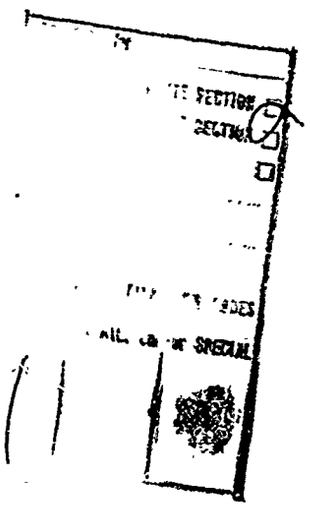

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# IDENTIFICATION AND YIELD OF Pd117 IN THE THERMAL-NEUTRON FISSION OF U235

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## ABSTRACT

As part of a study of nuclear charge distribution in the region of symmetric fission, the existence and cumulative fission yield of $^{117}Pd$ were sought. Palladium was isolated free from silver and cadmium at different times after fission and the quantity of descendant $^{117}Cd$ activity produced was measured. A half-life for $^{117}Pd$ of $5.0^{+0.5}_{-0.7}$ sec was derived from these data. Through a comparison of $^{117}Cd$ activity produced by $^{117}Pd$ with the total $^{117}Cd$ activity produced by all isobars, a cumulative fractional chain yield of 0.682 was computed for $^{117}Pd$. From this yield the most probable nuclear charge ($Z_p$) for the 117-mass chain was estimated.

## SUMMARY

### The Problem

As part of a general study of nuclear charge distribution in the region of symmetric fission of uranium, a search for a palladium isotope (mass 117) was conducted. This work represents an extension of the effort at NRDL to obtain data on the fission process for possible applications to fallout and nuclear reactor problems.

### Findings

$^{117}$Pd was identified and its half-life and cumulative fission yield were determined. An estimate of the most probable nuclear charge for mass chain 117 was derived.

## I. INTRODUCTION

$^{117}$Pd was identified and its cumulative fractional chain yield measured as an extension of a study on the distribution of nuclear charge in symmetric fission of $^{235}$U.

Palladium was separated from its descendants, silver and cadmium, at various intervals after irradiation by a procedure previously described.[1] The quantity of descendant $^{117}$Cd was measured in the palladium sample isolated. A plot of $^{117}$Cd activity as a function of time represented the radioactive decay of $^{117}$Pd. From this information, together with a measurement of the total $^{117}$Cd yield, the cumulative fractional yield of $^{117}$Pd was derived.

## II. EXPERIMENTAL

### Determination of $^{117}$Pd Half-life

The uranium solutions used in the irradiations were prepared as follows: $^{235}$U metal (93.22 % isotopic enrichment) was dissolved with concentrated nitric acid, and the excess acid was removed by several evaporations with concentrated hydrobromic acid. The dried salt was finally dissolved in concentrated hydrobromic acid at a concentration of 400 mg uranium/ml. To 7.5 ml of this solution were added 1.0, 1.5 and

2.0 ml of ruthenium, rhodium, and palladium carriers, each in 3N hydrochloric acid and at concentrations of 10, 6.4 and 10.0 mg metal/ml, respectively. Also added was 1 ml of silver carrier (10 mg of Ag/ml) in concentrated hydrobromic acid.

In each of several runs, 1 ml of the uranium solution contained in a sample carrier (rabbit) was irradiated for 3 sec in the Vallecitos Nuclear Test Reactor in a thermal-neutron flux of about $10^{12}$ neutrons $sec^{-1}$ $cm^{-2}$. A gold foil taped to the rabbit was used to monitor the neutron flux, from which measurement and previous calibrations the number of fissions were calculated.[2]

After irradiation, the rabbit was pneumatically transferred (12 ft) to the laboratory in about 0.25 sec. By means of the automated programmed system previously described,[3] at various times after irradiation the fission solution was transferred from the rabbit through 2 g of copper powder which selectively removed palladium. (The powder was supported on a 10 μ stainless steel mesh, 7/8 in. in diameter.) This part of the chemical separation occurred in 0.64 sec. For removal of interfering contaminants, the copper powder was washed with about 5 ml of hot concentrated hydrobromic acid over the next 0.96 sec.

The copper powder was quantitatively dissolved with 12 ml of concentrated nitric acid and the solution was heated to evolve oxides of nitrogen. Two ml of standardized cadmium carrier (11.2 mg/ml) were added to the sample, which then was diluted to 100 ml with 3 N hydrochloric acid. A 50 ml aliquot was reserved for the palladium yield

determination; the remainder was analyzed radiochemically[3] for $^{117}Cd$ after a minimum delay of 8 minutes from the time of the initial palladium separation. The delay was introduced to allow for the full growth of $^{117}Cd$ from the 1.1 m $^{117}Ag$ daughter product.

The $^{117}Cd$ count rate was determined in a gas flow proportional counter with a 200 mg $cm^{-2}$ aluminum absorber. A semi-logarithmic plot of the counts versus time showed an initial decay attributable to 49 min. $^{118}Cd$ (from $\sim$ 2 sec $^{118}Pd$ precursor) followed by the decay of the $^{117}Cd$ isomers with an apparent half-life of 3.1 h.* The linear portion of the curve (3.1 h) was extrapolated back about 5 hours to the end of irradiation, and the count obtained was corrected for the cadmium[3] and palladium carrier yields and was normalized for $10^{11}$ fissions.

The palladium carrier yield in the initial separation was determined as follows: The 50 ml aliquot reserved for this determination was spiked with $^{109}Pd$ tracer and evaporated to dryness. The residue was dissolved in 0.4 N nitric acid, and palladium was extracted into chloroform after the addition of 1 ml 1 % dimethylglyoxime in ethyl alcohol. Palladium was back-extracted into concentrated ammonium hydroxide. The solution was sulfided with hydrogen sulfide gas and simultaneously acidified with hydrochloric acid until a precipitate formed. The palladium sulfide was collected and then dissolved in aqua regia; the solution was evaporated to dryness and then dissolved and diluted to a definite

*The beta radiations of $Cd^{115}$ are undetectable through the aluminum absorber.

volume with $1.5 \times 10^{-3}$ N hydrochloric acid. With an aliquot of this solution the quantity of palladium was determined through comparison of its atomic absorptivity at 244.8 mμ with that of a palladium standard prepared in hydrochloric acid of the same acidity. With another aliquot, the $^{109}Pd$ recovery was determined by gamma-ray counting, and this value was used to correct for the palladium losses incurred in the chemical purification.

## Total $^{117}Cd$ Activity Yield Determination

Each of four rabbits which contained 200 mg $^{235}U$ in 1 ml of 6 N nitric acid was irradiated for 20 sec. Following irradiation, to a 0.3 to 0.5 ml aliquot were added 2 ml of cadmium carrier, and 3 ml of concentrated nitric acid in which were dissolved 500 mg of copper. The solution volume was increased to 50 ml with 3 N hydrochloric acid. Copper sulfide was precipitated a minimum of 8 min after the irradiation. Then cadmium sulfide, ferric hydroxide, and cadmium hydroxide were precipitated as described previously.[3] The cadmium hydroxide precipitate was dissolved in 10 ml of 2 N hydrochloric acid and the solution was passed through a Dowex A-2 resin. After the resin was washed with a small volume of 0.1 N hydrochloric acid, cadmium was eluted with 1.5 M sulfuric acid. The eluate was scavenged with ferric hydroxide and then cadmium hydroxide was precipitated. This precipitate was dissolved and reprecipitated as the sulfide[3] and counted as described above. The count rate

was corrected for decay from the time of irradiation and for the yield of cadmium carrier[3] and then was normalized for $10^{11}$ fissions.

## III. RESULTS AND DISCUSSION

The counting rate of $^{117}$Cd for twelve palladium separations made at various times after irradiation appears in Fig. 1. (The abscissa represents the difference in time between the end of irradiation and the start of palladium separation.) A linear relationship between the logarithm of the counting rate and the time of separation is evident. From a least squares analysis of this data, a half life of $5.0^{+0.5}_{-0.7}$ sec for $^{117}$Pd was computed.

For the calculation of the $^{117}$Pd cumulative fission yield, the midpoint between the initiation and completion of filtration of the fission solution through the copper bed was regarded as the separation time. A correction was made for the loss of $^{117}$Ag daughter product from the copper bed during the washing interval. By means of $^{109}$Pd tracer with its $^{109}$Ag daughter it was determined that 12 % of the silver formed in the beta decay process is removed when the deposited palladium is washed with hydrobromic acid as in the separation procedure.

The corrected $^{117}$Cd activity produced from $^{117}$Pd was compared to the total $^{117}$Cd activity and a fractional cumulative yield of 0.682 was computed for $^{117}$Pd. From this yield, and with the assumption that the distribution of nuclear charge is Gaussian, a value for the most probable

nuclear charge ($Z_p$) was estimated through the method of Wahl and co-workers.[4] In the absence of knowledge of the width parameter ($\sigma$) for the 117-mass chain, the estimation was made with the values 0.45 and 0.68, the range over which the width parameter was shown experimentally to vary.[5] The $Z_p$ derived from these extremes was 46.28 and 46.17, respectively. The broader width most closely approximates the value of 46.1 predicted by the equal charge displacement postulate.[6] The insensitivity of $Z_p$ to variation in $\sigma$ follows from the fact that the cumulative yield for $^{117}Pd$ is situated in the vicinity of that for $Z_p$.

A more refined characterization of $Z_p$ for the 117 mass chain would be possible if the nuclear charge dispersion were delineated for this chain. Accordingly, determination of the independent yields of $^{117}Ag$ and $^{117}Cd$ is being undertaken.

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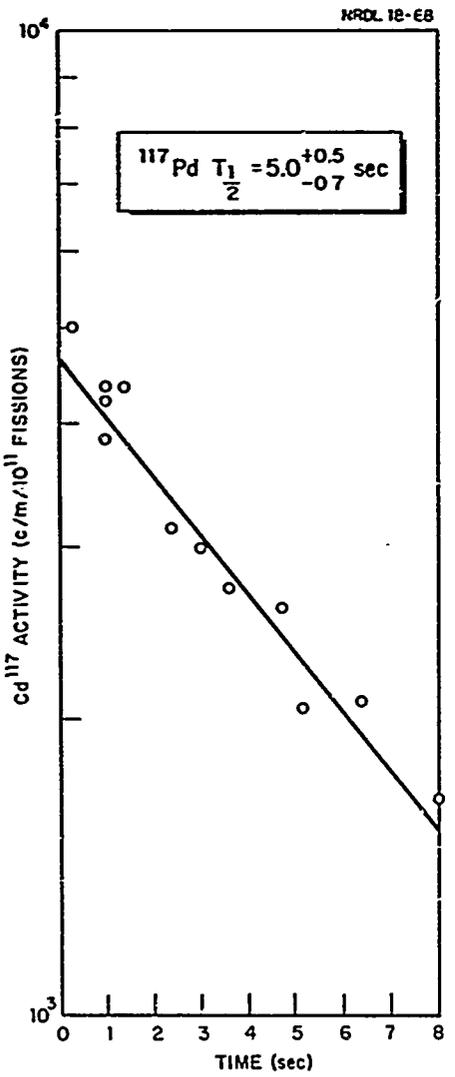


Fig. 1 The Growth of $^{117}$Cd Activity From Palladium Separated at Various Times After Fission.

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